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PolycaBonate SIABS

Polycarbonate sheets

The present invention relates to polycarbonate sheets, the use thereof to produce glazing and said glazing.

Polycarbonate sheets are used to produce glazing, for example. In this case, it is desirable to use sheets with as high as possible light transmissions and as low as possible yellowness indexes. Although it is possible to reduce the yellowness index by adding of blue pigments, this is achieved at the expense of light transmission and greying of the sheets. During weathering of the sheets, it is important that the mechanical properties of the sheets are kept at the highest possible level. Even after

weathering, the sheets must retain a high resistance to mechanical influences e.g.

hail.

15 Polycarbonates may be produced by the so-called interfacial polycondensation process. In said process, dihydroxydiarylalkanes (also known as bisphenols) are reacted in the form of their alkali salts in aqueous solution with phosgene in the heterogeneous phase in the presence of inorganic bases such as sodium hydroxide solution and an organic solvent, in which the product polycarbonate is readily soluble.

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More details relating to the interfacial polycondensation process for producing polycarbonates are disclosed in Schnell, "Chemistry and Physics of Polycarbonates", Polymer Reviews, Volume 9, Interscience Publishers, New York, London, Sydney

25 1964.

> The polycarbonate sheets known from the prior art exhibit the disadvantage that they do not adequately meet the above-mentioned requirements of high light transmission, low yellowness index and good retention of mechanical properties on weathering.

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 $\varphi_i(z; t_i^{(i)})$

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The object of the present invention is therefore to produce polycarbonate sheets which do not exhibit the above-mentioned disadvantages of the prior art.

This object is achieved according to the invention by polycarbonate sheets, wherein the polycarbonate is produced by the interfacial polycondensation process and the aqueous solution of an alkali salt of a bisphenol used therein has a content of dissolved oxygen of less than 150 ppb.

The content of dissolved oxygen in the aqueous solution of the alkali salt of bisphenol is <150 ppb, preferably <100 ppb, particularly preferably <50 ppb.

The invention thus provides polycarbonate sheets, wherein the polycarbonate is produced by the interfacial polycondensation process and the aqueous solution of an alkali salt of a bisphenol used therein has a content of dissolved oxygen of less than 150 ppb.

In addition, the present invention also provides glazing, for example for automobiles or buildings, in particular for greenhouses, winter gardens, bus shelters, advertising panels, signboards, protective screens, automotive glazing, windows or roofing.

The polycarbonates are produced according to the invention in that the other raw materials apart from the aqueous solution of the alkali salt of bisphenol do not contain a noteworthy amount of oxygen. In addition, production is performed with the exclusion of oxygen, as described for example in DE-A 4 227 272.

For example, in a reaction vessel and tubular reactor configuration, the forced circulation loop and the tubular reactors are flooded and the reaction vessel is blanketed with nitrogen, to ensure the exclusion of oxygen.

The polycarbonate sheets according to the invention are, for example, solid sheets, hollow sheets (also known as multi wall sheets e.g. twin wall sheets, triple wall

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sheets...) or corrugated sheets, which are e.g. used as glazing materials. The sheets may be produced in a manner known in principle to the person skilled in the art, for example by extrusion or by injection moulding.

Subsequent processing of the sheets, such as thermoforming or surface modification, such as application of scratch-proof lacquers, water-dispersing layers and the like, is also possible. The patent also relates to the moulded articles produced by these processes.

To produce the polycarbonate for the sheets according to the invention, aqueous solutions of an alkali salt of a bisphenol with a content of dissolved oxygen <150 ppb, preferably <100 ppb, particularly preferably <50 ppb are used, which may preferably be obtained by reacting bisphenols with a dissolved oxygen content <10 ppb with an aqueous alkali hydroxide solution with a dissolved oxygen content <100 ppb under oxygen exclusion.

Preferred alkali salts are the sodium salts of bisphenols.

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Bisphenols which may be used according to the invention are those which may be obtained by reacting ketones with aromatic hydroxy compounds which are not substituted in the para position and do not contain any second order substituents, such as cyano, carboxy or nitro groups, for example phenol, o- and m-cresol, 2,6-dimethylphenol, o-tert.-butylphenol, 2-methyl-6-tert.-butylphenol, o-cyclohexylphenol, o-phenylphenol, o-isopropylphenol, 2-methyl-6-cyclopentylphenol, o- and m-cresol, 2,6-dimethylphenol, o-tert.-butylphenol, preferably phenol, o- and m-cresol, 2,6-dimethylphenol, o-tert.-butylphenol and o-phenylphenol. Phenol is particularly preferred. Ketones with at least one aliphatic group on the carbonyl function are preferred, for example acetone, methyl ethyl ketone, methyl propyl ketone, methyl isopropyl ketone, diethyl ketone, acetophenone, cyclohexanone, cyclopentanone, methyl-, dimethyl- and trimethylcyclohexanones, which may also comprise geminal methyl groups, e.g. 3,3-dimethyl-5-methylcyclohexanone (hydroisophorone).

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Acetone, acetophenone, cyclohexanone and the homologues thereof bearing methyl groups are very particularly preferred. Acetone is the most preferable. By providing an intensive inert nitrogen atmosphere throughout the production process, it is ensured that the residual content of dissolved oxygen in the bisphenols is less than 10 ppb.

5 10 ppb

Bisphenols which may be used according to the invention are, additionally: 3-(4-hydroxyphenyl)-1,1,3-trimethylindan-5-ol; 1,3-di-(2-(4-hydroxyphenyl)-2-propyl)-benzene and 1,4-di-(2-(4-hydroxyphenyl)-2-propyl)benzene.

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Bisphenols which are particularly preferred are 2,2-bis(4-hydroxyphenyl)propane (i.e. bisphenol A) and 1,1-bis(4-hydroxyphenyl)-3,3,5-trimethylcyclohexane.

The bisphenols or the mixtures thereof are preferably reacted, with the exclusion of oxygen (e.g. by providing an inert nitrogen atmosphere), with aqueous alkali hydroxide solution having a dissolved oxygen content <100 ppb, preferably <20 ppb. The concentration of aqueous alkali hydroxide solution is preferably such that the concentration of the resultant aqueous solutions of an alkali salt of a bisphenol is as close as possible to the solubility limit, i.e. in the range of from 15 to 20 wt.%, preferably 16.5 to 18.5 wt.%. The molar ratio of alkali hydroxide to bisphenol is in particular from 1.8:1 to 2.5:1, preferably 1.9:1 to 2.4:1, particularly preferably 2.0:1 to 2.3:1. The bisphenol may be dissolved as a solid in aqueous alkali hydroxide solution. However, it is preferable for it to be added to the aqueous alkali hydroxide solution directly as a melt at temperatures of from 20°C to 90°C, preferably 30°C to 70°C, without its having to pass through the solid state.

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The virtually oxygen-free aqueous alkali hydroxide solution used to produce the aqueous solutions of an alkali salt of a bisphenol may be produced by electrolysis. After production, the aqueous alkali hydroxide solution should be stored and transported under inert gas. For use in the process according to the invention, the concentration of aqueous alkali hydroxide solution obtained during electrolysis is

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generally reduced by dilution with virtually oxygen-free, fully deionised water. Oxygen is removed from the fully deionised water in a manner known in principle, e.g. catalytically, by degassing or by inert gas stripping.

The aqueous solutions thus obtained of an alkali salt of a bisphenol exhibit particularly low colour indexes, which are naturally also dependent on the colour index of the bisphenol used. If a bisphenol is used which has a colour index <10 Hazen (measured according to ASTM D 1686), colour indexes of <1.5 Hazen, preferably <1.0 Hazen may be achieved.

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The polycarbonates may be branched deliberately and in a controlled manner by the use of small amounts of branching agent. Examples of suitable branching agents are: phloroglucinol, 4,6-dimethyl-2,4,6-tri-(4-hydroxyphenyl)-2-heptene; 4,6-dimethyl-2,4,6-tri-(4-hydroxyphenyl)heptane; 1,3,5-tri-(4-hydroxyphenyl)benzene; 1,1,1-tri-(4-hydroxyphenyl)ethane; tri-(4-hydroxyphenyl)phenylmethane; 2,2-bis-[4,4-bis(4-hydroxyphenyl)cyclohexyl]propane; 2,4-bis(4-hydroxyphenylisopropyl)phenol; 2,6-bis(2-hydroxy-5'-methylbenzyl)-4-methylphenol; 2-(4-hydroxyphenyl)-2-(2,4-dihydroxyphenyl)propane; hexa-(4-(4-hydroxyphenylisopropyl)phenyl)orthoterephthalic acid ester; tetra-(4-hydroxyphenyl)methane; tetra-(4-(4-hydroxyphenylisopropyl)phenoxy)methane); α,α',α'' -tris-(4-hydroxyphenyl)-1,3,5-triisopropylbenzene; 2,4-dihydroxybenzoic acid; trimesic acid; cyanuric chloride; 3,3-bis(3-methyl-4-hydroxyphenyl)-2-oxo-2,3-dihydroindole; 1,4-bis(4',4"-dihydroxytriphenyl)methyl)-benzene. 1,1,1-tri-(4-hydroxyphenyl)ethane and bis-(3-methyl-4-hydroxyphenyl)-2-oxo-2,3-dihydroindole (= isatinbiscresol).

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The branching agents or mixtures of branching agents which may optionally also be used, preferably in an amount of 0.05 to 2 mol-% relative to the bisphenols used, may be introduced together with the bisphenols or added at a later stage of the synthesis.

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Chain terminators may be used according to the invention. The chain terminators used according to the invention are preferably phenols such as phenol, alkylphenols such as cresol and 4-tert.-butylphenol, chlorophenol, bromophenol, cumylphenol or mixtures thereof. Phenol, 4-tert.-butylphenol or cumylphenol are particularly preferred.

Chain terminators and branching agents may be added to the reaction mixture as a separate solution or together with the bisphenolate, wherein care must be taken to ensure that the solutions have the same low oxygen content according to the invention as the aqueous solutions of an alkali salt of a bisphenol.

Other additives which may be used according to the invention and which may be added to the polycarbonate from which the sheets according to the invention are produced, are mentioned in EP-A 0 500 496 and WO 96/15102. According to the invention, mixtures of these additives may also be introduced into the polycarbonate.

Additives which are particularly preferred are UV absorbers such as the compounds described in WO 99/05205 of formula (I)

$$(R_1)_n$$

$$(R_1)_n$$

$$(R_2)_m$$

$$(R_2)_m$$

$$(R_2)_m$$

$$(R_2)_m$$

$$(R_2)_m$$

in which

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R¹ and R² are the same or different and mean H, halogen, C₁-C₁₀ alkyl, C₅-C₁₀ cycloalkyl, C₇-C₁₃ aralkyl, C₆-C₁₄ aryl, -OR⁵ or -(CO)-O-R⁵, with R⁵ being H or C₁-C₄ alkyl,

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 R^3 and R^4 are also the same or different and mean H, C_1 - C_4 alkyl, C_6 cycloalkyl, benzyl or C_6 - C_{14} aryl,

m is 1, 2 or 3 and

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ij

n is 1, 2, 3 or 4,

and also those of formula (II)

$$(R_1)_n$$
 $(R_1)_n$
 $(R_1)_n$
 $(R_2)_n$
 $(R_2)_n$
 $(R_2)_n$
 $(R_2)_n$
 $(R_3)_n$

in which the bridge means

R¹, R², m and n have the meaning stated in relation to formula (I),

in which, moreover, p is a whole number from 0 to 3,

20 q is a whole number from 1 to 10,

Y is $-CH_2-CH_2$, $-(CH_2)_3$, $-(CH_2)_4$, $-(CH_2)_5$, $-(CH_2)_6$, or $CH(CH_3)-CH_2$

and

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R³ and R⁴ have the meaning stated in relation to formula (I).

Other suitable UV absorbers are those comprising substituted triazines, such as 2,4-bis(2,4-dimethylphenyl)-6-(2-hydroxy-4-n-octyloxyphenyl)-1,3,5-triazine (CYASORB® UV-1164) or 2-(4,6-diphenyl-1,3,5-triazin-2-yl)-5-(hexyl)oxyphenol (Tinuvin® 1577). The UV absorber which is particularly preferred is 2,2-methylene bis(4-(1,1,3,3-tetramethylbutyl)-6-(2H-benzotriazol-2-yl)phenol), which is sold commercially under the names Tinuvin® 360 or Adeka Stab® LA 31. The UV absorber Uvinul 3030, made by BASF AG and obtained in accordance with Example 1 of WO 96/15102, is also suitable, as are the UV absorbers given in EP 0 500 496 A1.

Other suitable UV absorbers are 5'-methyl-, 3',5'-di-tert.-butyl-, 5'-tert.-butyl-, 5'-(1,1,3,3-tetramethylbutyl)-, (= Tinuvin 329), 5-chloro-3',5'-di-tert.-butyl-, 5-chloro-3'-tert.-butyl-5'-methyl-, 3'-sec.-butyl-5'-tert.-butyl, 4'-octoxy-, 3',5'-di-tert.-amyl-, 3',5'-bis(\alpha,\alpha-dimethylbenzyl)-, mixture of 5-chloro-3'-tert.-butyl-5'-(2-octyloxycarbonylethyl)- and 5-chloro-3'-tert.-butyl-5'-[2-(2-ethylhexyloxy)carbonylethyl]-, 5chloro-3'-tert.-butyl-5'-(2-methoxycarbonylethyl)-, 3'-tert.-butyl-5'-(2-methoxycarbonylethyl)-, 3'-tert.-butyl-5'-(2-octyloxycarbonylethyl)-, 3'-tert.-butyl-5'-[2-(2-ethylhexyloxy)-carbonylethyl]-, 3'-dodecyl-5'-methyl- and 3'-tert.-butyl-5'-(2-isooctyloxycarbonylethyl)-2'-hydroxyphenyl-2H-benzotriazole(2), 2,2'-methylenebis[4-(1,1,3,3-tetramethylbutyl)-6-benzotriazol-2-yl-phenol] together with 3'-sec.-butyl-5'-(1,1,3,3-tetramethylbutyl-2'-hydroxyphenyl-2H-benzotriazole (= Tinuvin 350) and 2-[2'-hydroxy-3',5'-bis(1,1-dimethylbenzyl)phenyl]-2H-benzotriazole (= Tinuvin 234) and 2-hydroxybenzophenones, such as for example 4-hydroxy-, 4-methoxy-, 4octoxy-, 4-decyloxy-, 4-dodecyloxy-, 4-benzyloxy-, 4,2',4-trihydroxy-, 2'-hydroxy-4,4'-dimethoxy derivative, ethyl-2-cyano-3,3-diphenyl acrylate (= Uvinul 3035) made by BASF AG, 2-ethylhexyl-2-cyano-3,3-diphenyl acrylate (= Uvinul 3039) made by BASF AG.

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Examples of suitable stabilisers, which may be used as additives according to the invention, are phosphines, phosphites or Si-containing stabilisers and other compounds described in EP-A 0 500 496. Triphenyl phosphites, diphenylalkyl phosphites, phenyldialkyl phosphites, tris(nonylphenyl) phosphite, triphenyl phosphine, tetrakis(2,4-di-tert.-butylphenyl)-4,4'-biphenylene diphosphonite and triarylphosphite may be cited by way of example. Triphenylphosphine and tris(2,4-di-tert.-butylphenyl)phosphite are particularly preferred.

The polycarbonates, from which the sheets according to the invention are produced, may additionally contain additives to improve the surface slip characteristics, for example esters of polyhydric alcohols with long-chain fatty acids, such as for example pentaerythritol tetrastearate/-palmitate, glycerol monostearate/-palmitate, glycerol tristearate/-palmitate. Furthermore, the addition of flame retardants, pigments, colorants, finely divided minerals, optical brighteners and other additives is also possible.

All the feed materials and solvents used for synthesis may be contaminated with impurities from the production and storage thereof, wherein the aim is to work with starting materials which are as clean as possible.

Glazing according to the invention comprises, in particular, glazing for automobiles or buildings, for example glazing for greenhouses, winter gardens, bus shelters, advertising panels, signboards, protective screens, automotive glazing, windows or roofing.

Subsequent processing of the sheets according to the invention, such as thermoforming or surface modification, such as application of scratch-proof lacquers, water-dispersing layers and the like, is possible.

Examples

In the following Examples, the colour indexes were determined according to ASTM D 1686 by measuring absorption up to 400 nm over an irradiation length of 50 cm.

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Weathering of these sheets (according to ISO 4892-2A) was performed in a Weatherometer made by Atlas, USA, using a 6.5 W xenon lamp and a cycle involving 102 mins of exposure to light and 18 mins of spraying with deionised water together with light exposure. The maximum black body temperature was 60°C (± 5°C). After 1000 hours, samples of the sheets were taken and toughness was tested under practical conditions using a modified falling dart test which was developed for multi-walled sheets on the basis of DIN 53 443. The fracture energy W_s being measured using a 5 mm diameter puncture body intended to cause biaxial stress similar to that caused by a falling hailstone. At room temperature, a falling mass of 36 kg falling from a height of 0.2 m struck the samples, which lay freely on a supporting ring with an internal diameter of 20 mm.

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Testing was performed with the weathered side of the sample in the compression zone. The fracture appearance (BB) was characterised by numbers: 1 = splintered, 2 = smooth crack, 3 = ductile/brittle, 4 = ductile.

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The yellowness index determined according to ASTM E 313 was stated as Yellowness Index YI.

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Example 1

To produce 1.022 t/h of a 15% aqueous NaBPA solution, 867.5 kg/h of a 6.5% aqueous sodium hydroxide solution and 154.5 kg/h of bisphenol A melt were combined continuously. The entire process was performed under an inert atmosphere of nitrogen. The 6.5% aqueous sodium hydroxide solution comprised an oxygen content of 10 ppb. The Hazen colour index of the resultant 15% aqueous sodium bisphenolate solution amounted to 0.5 Hazen. This sodium bisphenolate solution was used to produce polycarbonate by the interfacial polycondensation process.

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Comparative Example 2

To produce 1.022 t/h of a 15 % aqueous sodium bisphenolate solution, 867.5 kg/h of a 6.5% aqueous sodium hydroxide solution and 154.5 kg/h of bisphenol A melt were combined continuously. The entire process was performed under an inert atmosphere of nitrogen. The 6.5% aqueous sodium hydroxide solution comprised an oxygen content of 250 ppb. The Hazen colour index of the resultant 15% aqueous sodium bisphenolate solution amounted to 2 Hazen. This sodium bisphenolate solution was used to produce polycarbonate by the interfacial polycondensation process.

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Example 3

A polycarbonate with phenol end groups, a melt flow index (MFR) of 3 (measured according to ISO 1133), a branching agent content of 0.3 mol% isatinbiscresol and a UV absorber content of 0.25% Tinuvin® 350 (3'-sec.-butyl-5'-(1,1,3,3-tetramethyl-butyl-2'-hydroxyphenyl-2H-benzotriazole) was produced from the sodium bisphenolate solution obtained in Example 1. This polycarbonate was processed in the following plant to produce 10 mm coextruded twin-wall sheets:

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main extruder having a screw of length 33D and a diameter of 70 mm with venting

- coextruder for applying the outer layer having a screw of length 25D and a diameter of 30 mm
- special sheet die with a width of 350 mm
- calibrator
- 5 roller conveyor

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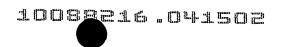
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- take-off unit
- cutting device (saw)
- stacking table.
- The polycarbonate obtained from Example 1 was fed introduced into the coextruder.

 Coextrusion as such is known in the art (see for example EP-A 110 221 and EP-A 110 238). In the present instance, the process was performed as follows:
 - Extruders for producing the core layer and the outer layer(s) were connected to a coextrusion adapter. The adapter was designed that the melt forming the outer layer was applied as a thin layer adhering to the core layer melt.
 - The multi-layer melt strand thereby produced was then given the desired form (twin-walled sheet) in the die connected downstream. The melt was then cooled in a known way under controlled conditions by means of vacuum calibration and subsequently sawn into pieces 1 metre in length.

Comparative Example 4

A polycarbonate with phenol end groups, a melt flow index (MFR) of 3 (measured according to ISO 1133), a branching agent content of 0.3 mol% isatinbiscresol and a UV absorber content of 0.25% Tinuvin® 350 (3'-sec.-butyl-5'-(1,1,3,3-tetramethyl-butyl-2'-hydroxyphenyl-2H-benzotriazole) was produced from the sodium bisphenolate solution obtained in Example 2. This polycarbonate was processed in the same plant and under the same conditions as in Example 3 to produce 10 mm coextruded twin-wall sheets.



Example 5

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A polycarbonate with phenol end groups and a melt flow index (MFR) of 3 (according to ISO 1133) was produced from the sodium bisphenolate solution obtained in Example 1. This polycarbonate was processed in the same plant and under the same conditions as in Example 3 to produce 10 mm coextruded twin-wall sheets.

A compound of the polycarbonate obtained from Example 1 and 5% of a benzotriazole UV absorber (Tinuvin® 360 = 2,2-methylenebis-(4-(1,1,3,3-tetramethylbutyl)-6-(2H-benzotriazol-2-yl)phenol) was introduced into the coextruder. This compound was produced in the following manner: the UV absorber was mixed into the polycarbonate in a twin-screw extruder (ZSK 32, Werner & Pfleiderer) at 280°C and 80 rev/min and the extrudate was then pelletised.

Comparative Example 6

A polycarbonate with phenol end groups and a melt flow index (MFR) of 3 (according to ISO 1133) was produced from the sodium bisphenolate solution obtained in Example 2. This polycarbonate was processed in the same plant and under the same conditions as in Example 3 to produce 10 mm coextruded double-wall sheets.

A compound of the polycarbonate obtained from Example 1 and 5% of a benzo-triazole UV absorber (Tinuvin® 360 = 2,2-methylenebis-(4-(1,1,3,3-tetramethyl-butyl)-6-(2H-benzotriazol-2-yl)phenol) was introduced into the coextruder. This compound was produced in the following manner: the UV absorber was mixed into the polycarbonate in a double-shaft extruder (ZSK 32, Werner & Pfleiderer) at 280°C and 80 rev/min and the extrudate was then pelletised.

Example 7

A polycarbonate with phenol end groups, a melt flow index (MFR) of 3 (measured according to ISO 1133) and a UV absorber content of 0.3% Tinuvin® 350 (3'-sec.-butyl-5'-(1,1,3,3-tetramethylbutyl-2'-hydroxyphenyl-2H-benzotriazole) was produced from the sodium bisphenolate solution obtained in Example 1. This polycarbonate was processed by injection moulding to produce colour chips 3 mm thick.

Comparative Example 8

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A polycarbonate with phenol end groups, a melt flow index (MFR) of 3 (measured according to ISO 1133) and a UV absorber content of 0.3% Tinuvin® 350 (3'-sec.-butyl-5'-(1,1,3,3-tetramethylbutyl-2'-hydroxyphenyl-2H-benzotriazole) was produced from the sodium bisphenolate solution obtained in Example 2. This polycarbonate was processed by injection moulding to produce colour chips 3 mm thick.

The sheets obtained were then subjected to colorimetric assessment, using the following measuring methods:

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- 1. Transmission (on the basis of standard ASTM D 1003)

 Apparatus: Pye-Unicam (measuring geometry: 0°/diffuse, calculated according to illuminant C)
 - 2. Yellowness index YI according to ASTM E 313

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3. Weathering of these sheets was performed according to ISO 4892-2A in the Weatherometer made by Atlas, USA using a 6.5 W xenon lamp and a cycle involving 102 mins of exposure to light and 18 mins of spraying with deionised water together with light exposure. The maximum black body temperature was 60°C (± 5°C). After 1000 hours, samples of the sheets were taken and toughness was tested under practical conditions using a modified



falling dart test developed for multi wall sheets on the basis of DIN 53 443, the fracture energy W_s being measured using a 5 mm diameter puncture body intended to cause biaxial stress similar to that caused by a falling hailstone. At room temperature, a falling mass of 36 kg falling from a height of 0.2 m struck the samples, which lay freely on a supporting ring with an internal diameter of 20 mm.

Testing was performed with the weathered side of the sample in the compression zone. The fracture appearance (BB) was characterised by numbers: 1 = splintered, 2 = smooth crack, 3 = ductile/brittle, 4 = ductile.

The yellowness index determined according to ASTM E 313 was stated as Yellowness Index YI.

15 Results:

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Transmission (%)	Yellowness Index
79.5	1.2
77.4	2.7
87.3	3.0
84.0	5.5
	79.5 77.4 87.3

	Appearance of fracture (0 hours weathering as stated)	Appearance of fracture (4000 hours weathering as stated)	Appearance of fracture (5000 hours weathering as stated)
Example 3	4x4	4x4	4x4
Comparative example 4	4x4	4x3	4x3
Example 5	4x4	4x4	4x4
Comparative example 6	4x4	3x4, 1x3	3x4, 1x3

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The results show that sheets produced according to the invention have higher transmission values together with lower Yellowness Index values and better mechanical stability after weathering than the sheets according to comparative tests.